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Polymerization shrinkage and shrinkage force kinetics of high- and low-viscosity dimethacrylate- and ormocer-based bulk-fill resin composites

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Keywords:

Bulk-fill composite • Ormocer • Degree of conversion • Polymerization contraction • Shrinkage stress

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Abstract

The aim of the present study was to investigate polymerization shrinkage, shrinkage force development, and degree of monomer conversion of high- and low-viscosity dimethacrylate- and ormocer-based bulk-fill resin composites. Two flowable bulk-fill composites (SDR, x-tra base), two high-viscosity bulk-fill composites (Bulk Ormocer, SonicFill), and two conventional composite materials (Esthet X flow, Esthet X HD) were photoactivated for 20 s at 1,275 mW/cm^2. Linear polymerization shrinkage and shrinkage force were recorded in real time using custom-made devices, and the force rate and time to achieve maximum force rate were determined. Degree of conversion was measured using Fourier transform infrared spectroscopy. Data were analyzed with one-way ANOVA and Tukey's HSD post-hoc test, and bivariate correlations were computed (α = 0.05). The category of high-viscosity bulk-fill resin composites showed the significantly lowest polymerization shrinkage and force development. Within the tested flowable composite materials, SDR bulk-fill generated the significantly lowest shrinkage forces during polymerization and attained the significantly highest degree of conversion. Strong positive correlations were revealed between shrinkage force and both linear polymerization shrinkage (r = 0.902) and maximum force rate (r = 0.701). Linear shrinkage and shrinkage force both showed a negative correlation with filler volume content (r = -0.832 and r = -0.704, respectively). Bulk-fill resin composites develop lower shrinkage forces than their conventional flowable and high-viscosity counterparts, respectively, which supports their use for restoring high C-factor posterior cavities. Overall, bulk-fill composites with high filler amount and low force rate showed the most favorable shrinkage force characteristics.

Introduction

In an attempt to simplify and expedite the placement of direct composite restorations, so-called bulk-fill resin composites have been introduced to the dental profession. These materials are characterized by an increased depth of cure compared with conventional resin composites, allowing thick layers of up to 4–5 mm to be placed and photo-polymerized in a single step [1–4]. Besides saving clinical time, bulk placement can prevent incorporation of voids or contaminants between composite layers [5–7]. Bulk-fill resin composites vary in their rheological properties and are available as low-viscosity (flowable) and high-viscosity (sculptable) materials. The inferior mechanical properties of the former require to finish the restoration by applying a capping layer made of a regular composite material, whereas high-viscosity bulk-fill composites are indicated for use without veneering, but are more difficult to adapt to cavity walls due to their high filler load [8, 9].

One of the main concerns when bulk-curing large volumes of resin-based composite materials is polymerization shrinkage stress emerging at the tooth-restoration interface. Shrinkage stress is produced by polymerization contraction of a resin composite under conditions of constraint, created by bonding to cavity walls, and has been associated with a series of clinical complications including cuspal deflection, interfacial debonding, post-operative sensitivity, and secondary caries [10, 11]. However, shrinkage stress not only depends on the composite's volumetric shrinkage, which is proportional to the degree of monomer to polymer conversion [12, 13]. It is, beyond that, also largely determined by the time-dependent visco-elastic properties of the material, defined by flow capacity in early stages of the polymerization reaction and elastic modulus development during polymer network formation [12, 14].

In order to control reaction kinetics and minimize stress formation in composite restorations, manufactures incorporated proprietary high-molecular-weight base monomers, pre-polymer stress relievers, and stress-relaxant polymerization modulators in their bulk-fill materials [15, 16]. While previous *in vitro* research substantiated that bulk-fill resin

composites cause lower contraction forces [17, 18] and less cuspal flexure [19, 20] than conventional composite materials, other studies revealed no advantage of bulk-fill over conventional nanohybrid composites in terms of shrinkage stress development [21] and restoration margin integrity [22, 23]. Furthermore, conflicting data exists on whether high- or low-viscosity bulk-fill resin composites display favorable shrinkage stress kinetics [24, 25]. While the addition of inorganic fillers to the resin matrix decreases overall shrinkage of the composite material [26, 27], it concurrently increases its elastic modulus [28]. Which effect plays the larger role in shrinkage stress development is still a topic of debate.

Besides the inorganic filler content, the resin matrix has an important influence on the shrinkage behavior of dental composite materials [29]. Other than traditional monomer systems containing dimethacrylates such as Bis-GMA, UDMA and TEGDMA, ormocer-matrix-based composites are inorganic-organic hybrid polymers that form, by hydrolysis and condensation of alkoxides, an inorganic Si–O–Si network selectively modified through the incorporation of covalently-bonded organic groups [30–32]. Due to their reduced amount of organic resin, ormocers have been reported to cause lower polymerization contraction compared with dimethacrylate-based composite materials [33, 34]. However, scientific data on the effect of ormocer technology on shrinkage force formation is scarce and inconsistent [35, 36]. Moreover, up to date, no information is available in the literature on shrinkage characteristics of bulk-fill composites containing ormocer-based resin matrices.

Therefore, the aim of the present study was to investigate the polymerization shrinkage, the shrinkage force kinetics and degree of conversion of high- and low-viscosity dimethacrylate- and ormocer-based bulk-fill resin composites. The null hypothesis was that there would be no differences in the polymerization shrinkage behavior and force development between bulk-fill and conventional resin composite materials.

Materials and methods

Six resin composite materials were tested in this study: two flowable bulk-fill composites [SDR (Dentsply DeTrey, Konstanz, Germany) and x-tra base (VOCO, Cuxhaven, Germany)], two high-viscosity bulk-fill composites [Bulk Ormocer (VOCO) and SonicFill (Kerr, Orange, CA, USA)], a conventional flowable composite [Esthet X flow (Dentsply DeTrey)], and a conventional high-viscosity composite [Esthet X HD (Dentsply DeTrey)]. Details of the test materials are given in Table 1. In all tests, photoactivation was performed using an LED light curing unit (Bluephase G2; Ivoclar Vivadent, Schaan, Liechtenstein) with a 10-mm diameter light emission window. Output irradiance (1,275 mW/cm^2) of the curing light was measured with a calibrated FieldMaxII-TO power meter in conjunction with a PM2 thermopile sensor (Coherent, Santa Clara, CA, USA), and checked periodically during the study. Experimental testing was conducted inside a temperature-controlled chamber at 25°C [21], which simulates intraoral temperature conditions after rubber dam placement [37].

Linear shrinkage

Linear shrinkage was determined using a custom-made linometer (Fig. 1), as previously described in the literature [38–40]. In brief, a standardized amount (42 mm^3) of composite material was applied on a thin aluminum platelet, which was loosely placed upon the solid metal frame of the linometer. The underside of the platelet featured a perpendicular diaphragm, which extended into a recess of the linometer's infrared measuring sensor. The composite material to be tested was flattened to a thickness of 1.5 mm by means of a sandblasted (aluminum oxide, 50 µm; Renfert, Hilzingen, Germany) and silanized (Monobond Plus; Ivoclar Vivadent) glass plate. Prior to silanization, aluminum oxide particles were removed from the glass plate by washing the glass plate with an air water spray for 30 s. After air-drying, no embedded aluminum oxide particles could be observed in the glass plate with a stereomicroscope at 40x magnification (M3Z; Leica/Wild, Heerbrugg, Switzerland). Photoactivation of the specimens was performed for 20 s through the glass plate. The vertical displacement of the diaphragm induced by polymerization shrinkage of the

composite materials was detected by the infrared sensor at a sampling frequency of 5 Hz during 15 min from the start of photoactivation. Five specimens were tested for each material.

Shrinkage force

Measurements of polymerization shrinkage force were carried out using a custom-made stress analyzer (Fig. 2), based on principles as also described in detail previously [38–40]. Briefly, the device comprised a semi-rigid load cell (PM 11-K; Mettler, Greifensee, Switzerland; instrument compliance: 0.4 µm/N), to which a metal cylinder was connected. Composite material (42 mm^3) was applied to the front side of the cylinder and compressed to a thickness of 1.5 mm by a glass plate, which was fixed to the under side of the device. In order to improve adhesion, the surfaces of the metal cylinder and the glass plate were sandblasted with aluminum oxide (50 µm; Renfert), subsequently cleaned as described above, and coated with a universal primer (Monobond Plus; Ivoclar Vivadent). Photoactivation of the composite specimens was performed for 20 s through the glass plate, and the forces produced during polymerization shrinkage were detected by the load cell at a 5-Hz sampling frequency. Measurements (n = 5 per material) were carried out for 15 min from the start of photoactivation. The force rate was calculated as the first derivative of the shrinkage force vs. time curve, and the kinetic parameters maximum force rate (R_{max}) and time to achieve maximum force rate (t_{Rmax}) were determined.

Degree of conversion

Degree of conversion (DC) was measured using a Fourier transform infrared spectrometer (Cary 630 FTIR; Agilent Technologies, Santa Clara, CA, USA) with an attenuated total reflectance (ATR) accessory [41]. The same amount of composite (42 mm^3) as for assessment of linear shrinkage and shrinkage force was applied on the ATR diamond crystal with the aid of a 1.5 mm thick cylindrical Teflon mold, which was covered with a sandblasted (aluminum oxide, 50 µm; Renfert) and silanized (Monobond Plus; Ivoclar Vivadent) glass

plate. Prior to silanization, the glass plate was cleaned as described above. Photoactivation of the resin composite materials was conducted for 20 s through the glass plate. Infrared spectra of both the unpolymerized and polymerized composite materials were recorded in absorbance mode in the 4000–400 cm^{-1} wave number range at a resolution of 4 cm^{-1}. The absorbance intensities (peak heights) of the aliphatic C=C stretching vibrations at 1637 cm^{-1}, and aromatic C · · ·C stretching vibrations (internal standard) at 1608 cm^{-1} (for the dimethacrylate-based composites) or 1592 cm^{-1} (for the ormocer-based composite) were determined by using a baseline method [42]. The degree of conversion at 15 min after the start of photoactivation (endpoint of observation period for linear shrinkage and shrinkage force measurements) was calculated according to the equation [43]:

$$DC\ (\%) = \left(1 - \frac{R_{cured}}{R_{uncured}}\right) \times 100$$

where R is the ratio of absorbance intensities of the 1637 cm^{-1} and 1608 cm^{-1} peaks in the spectra of the dimethacrylate-based composites, or the 1637 cm^{-1} and 1592 cm^{-1} peaks in the spectra of the ormocer-based composite. Five specimens were tested per material.

Statistical analysis

ANOVA assumptions (normality and homogeneity of variance) were checked by means of the Kolmogorov-Smirnov and Levene tests. One-way ANOVA followed by Tukey's HSD post-hoc test was performed to identify differences in means of continuous variables between materials. In addition, bivariate correlations were computed. All statistical analyses were conducted at a pre-set level of significance of $\alpha = 0.05$ (SPSS Version 20; SPSS, Chicago, IL, USA).

Results

Figures 3 and 4 show the time-dependent development of linear shrinkage and shrinkage force curves, respectively. The linear shrinkage and shrinkage force values recorded at the end of the 15-min observation period, as well as the maximum force rate (R_{max}), the time to achieve maximum force rate (t_{Rmax}), and the degree of conversion of the investigated composite materials are presented in Table 2.

The high-viscosity composite materials under investigation (Esthet X HD, Bulk Ormocer, and SonicFill) developed significantly lower linear shrinkage than the low-viscosity resin composites (Esthet X flow, SDR, and x-tra base). Within the high-viscosity composites, the ormocer-based bulk-fill material (Bulk Ormocer) showed the significantly lowest linear shrinkage, followed by the dimethacrylate-based bulk-fill material (SonicFill) and the conventional composite (Esthet X HD). Within the low-viscosity materials, the bulk-fill composites (SDR and x-tra base) developed significantly lower linear shrinkage than the conventional composite (Esthet X flow) ($p < 0.001$ and $p < 0.001$, respectively).

The significantly lowest shrinkage force was generated by the ormocer-based bulk-fill composite (Bulk Ormocer). This material also showed the lowest maximum force rate (R_{max}), although the mean value was not statistically significantly different from R_{max} of SDR, Esthet X HD, and SonicFill. The dimethacrylate-based high-viscosity bulk-fill composite SonicFill developed significantly lower shrinkage force compared with both the flowable bulk-fill composite SDR ($p < 0.001$) and the conventional high-viscosity composite Esthet X HD ($p < 0.001$), which were both in the same statistical group. The dimethacrylate-based low-viscosity bulk-fill material x-tra base showed significantly higher shrinkage force than both SDR ($p = 0.001$) and Esthet X HD ($p = 0.001$), but significantly lower shrinkage force compared with the conventional flowable composite material (Esthet X flow) ($p < 0.001$).

The bulk-fill materials SonicFill and SDR achieved the significantly highest degrees of conversion among the tested resin composites. Bulk Ormocer, x-tra base, and Esthet X HD

were ranked in the same statistical group, and reached a significantly lower degree of conversion compared with Esthet X flow ($p = 0.047$, $p = 0.001$ and $p < 0.001$, respectively).

Strong positive correlation was found between linear shrinkage and shrinkage force ($r = 0.902$, $p < 0.001$). Moreover, a positive correlation was revealed between shrinkage force and maximum force rate (R_{max}) ($r = 0.701$, $p < 0.001$), but not between shrinkage force and degree of conversion ($p > 0.05$). Both linear shrinkage and shrinkage force showed a negative correlation with filler weight content ($r = -0.954$, $p < 0.001$ and $r = -0.870$, $p < 0.001$, respectively) and filler volume content ($r = -0.832$, $p < 0.001$ and $r = -0.704$, $p < 0.001$, respectively).

Discussion

The present study investigated linear polymerization shrinkage, the resulting shrinkage force kinetics, as well as the degree of monomer conversion of various bulk-fill and conventional composite materials. It revealed significantly lower shrinkage and shrinkage force development of the bulk-fill resin composites compared with their conventional flowable and high-viscosity counterparts, respectively, which led to the rejection of the null hypothesis. Overall, the category of high-viscosity bulk-fill resin composites showed the most favorable shrinkage force behavior at clinically acceptable levels of monomer conversion of > 55% [44].

The main reason identified for the low shrinkage force development of the dimethacrylate- and ormocer-based bulk-fill resin composites under investigation was their low polymerization shrinkage, and a strong positive correlation between linear shrinkage and shrinkage force ($r = 0.902$) was established in accordance with previous reports [24, 38]. In the current shrinkage tests, the use of a sandblasted glass plate might have caused a resistance to shrinkage at this surface, and increased the shrinkage in the axial direction. However, relative differences in the shrinkage behavior of the tested materials are not

affected by the type of glass plate used. The lower linear shrinkage of the tested bulk-fill resin composites compared with their conventional flowable and high-viscosity counterparts, respectively, might be explained by the higher filler content of the bulk-fill materials (Table 1). At a high filler content, the amount of organic matrix and, consequently, the number of reactive methacrylate groups, is reduced, resulting in lower polymerization shrinkage and thus lower shrinkage force development [45]. As a matter of fact, in the current investigation, both linear shrinkage and shrinkage force showed a strong negative correlation with filler content (by weight and by volume).

However, a higher filler content not only decreases polymerization shrinkage of resin-based composites, but simultaneously increases their elastic modulus [28], which, in turn, increases shrinkage stresses according to Hooke's law. Accordingly, other studies found a positive correlation between filler content and shrinkage stress [46, 47]. In these studies, however, in contrast to the present investigation, near-zero compliance (highly rigid) test set-ups were used, containing negative feedback mechanisms to fully compensate axial specimen deformation during stress measurements. While the elastic modulus has been revealed as the decisive factor affecting shrinkage stress formation in these highly rigid testing systems, shrinkage stress development depends more on polymerization shrinkage than on elastic modulus when more compliant (semi-rigid) measuring devices are used [48–50]. Consequently, conflicting results on the effect of inorganic filler content on shrinkage stress might be ascribed to differences in the compliance of measuring devices [51]. Since real teeth and their cavities are not completely rigid, but show elastic and visco-elastic characteristics [52], in the current experimental set-up, axial deformation of the specimens was only partially restricted, with the load cell being axially displaced by 0.4 µm per Newton force. According to Wang and Chiang [51], an instrument compliance of 0.4 µm/N falls in the category of low compliance, in which the stress should increase with the filler content. However, in the present study, shrinkage forces, in general, decrease with the increase of filler content (decrease of linear shrinkage). In other words, the trend observed in this study should fall into the category of high compliances. One may speculate, but literature suggests

that the materials do not have equivalent elastic moduli [53], so some other reason must be responsible for this behavior, due to the complex nature of stress development in these composite systems.

In the present investigation, the experimental ormocer-based bulk-fill resin composite (Bulk Ormocer) developed the significantly lowest linear polymerization shrinkage and shrinkage force. The low shrinkage of the ormocer matrix can be ascribed to its resin system consisting of inorganic–organic copolymers instead of classic monomers (e.g. Bis-GMA, UDMA and TEGDMA), and its reduced amount of organic resin compared with dimethacrylate-based composites [30, 31]. Besides the low polymerization shrinkage, the low maximum force rate (R_{max}) measured for Bulk Ormocer might have contributed to its favorable shrinkage force behavior. The low value for R_{max} indicates that Bulk Ormocer generates polymerization-induced forces at a slower rate, which gives the developing polymer network more time to re-arrange itself during the initial curing stage, and thereby to partially dissipate emerging shrinkage forces by viscous flow and polymer chain relaxation before mobility is restrained by vitrification [39, 54, 55]. Furthermore, the degree of conversion of Bulk Ormocer was much lower than that of the other high viscous bulk-fill composite studied, SonicFill, which can contribute to lower linear shrinkage and shrinkage force formation. Finally, the resin content in the ormocer composite was slightly lower than that in SonicFill, which can also contribute to the observed lower linear shrinkage and shrinkage force formation of the ormocer composite.

Within the low-viscosity composites, SDR bulk-fill generated the lowest linear shrinkage and shrinkage force, followed by the other flowable bulk-fill material x-tra base, even thought SDR contains a lower filler amount. The favorable shrinkage behavior of SDR might be explained by its unique resin composition, which contains a modified high-molecular-weight UDMA base monomer with reduced density of reactive sites per unit of mass and thus low polymerization shrinkage. Furthermore, a ‘polymerization modulator’ has been embedded as chemical moiety in the center of the polymerizable organic SDR matrix, enabling the monomers to react more flexibly and at a slower rate [56], which has

been shown to result in internal stress relaxation without compromising the degree of conversion [36, 40].

Although bulk-fill composites can be applied in up to 4–5 mm increment thickness clinically, in the current study, specimen thickness was set to 1.5 mm to be able to compare linear shrinkage and shrinkage force development of the bulk-fill materials with that of conventional resin composites. Conventional composites are only indicated for use in max. 2 mm thick increments. Therefore, these materials would not polymerize properly at 4–5 mm specimen thickness, in contrast to their bulk-fill counterparts [4]. Moreover, if the thickness of only the bulk-fill composites had been increased, the C-factor would have been simultaneously lowered for the bulk-fill composites. In the current test set-up, increased composite thickness would enable more radial stress relief by viscous flow from free, unbonded surfaces, and thus reduce the measurable axial stress [57]. One limitation of the force-measuring device used in the present study is that different filling techniques (application of bulk-fill composites in thick layers vs. application of conventional composites in thinner layers) cannot be adequately compared for the reason given above. In order to compare different filling techniques, cuspal deflection tests are well suited.

Within the limitations of the present *in vitro* study, it can be concluded that bulk-fill resin composites contract less during polymerization and develop lower shrinkage forces compared with their conventional flowable and high-viscosity counterparts, respectively. Overall, bulk-fill composites with reduced amount of organic matrix and low force rate showed the most favorable shrinkage force characteristics, which supports their use for restoring high C-factor posterior cavities.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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Table 1 Manufacturers' information about the resin composite materials used in the study

Material	Composition	Filler size (μm)	Filler content (wt%/vol%)	Lot no.	Manufacturer
Esthet X HD	Matrix: Bis-GMA adduct, Bis-EMA, TEGDMA Filler: Ba-F-Al-B-Si-glass, SiO_2	0.02–3.0 (mean: 0.61)	76/60	140829	Dentsply DeTrey, Konstanz, Germany
Esthet X flow	Matrix: Bis-GMA adduct, Bis-EMA, TEGDMA Filler: Ba-F-Al-B-Si-glass, SiO_2	0.02–7.5 (mean: 1.2)	61/53	110531	Dentsply DeTrey, Konstanz, Germany
Bulk Ormocer	Matrix: ORMOCER* matrix Filler: Glass ceramics, SiO_2	0.02–3.0	84/69	1441426	VOCO, Cuxhaven, Germany
SonicFill	Matrix: Bis-GMA, Bis-EMA, TEGDMA Filler: Ba-B-Al-Si-glass, SiO_2	Not indicated	83.5/66	5338301	Kerr, Orange, CA, USA
SDR	Matrix: Modified UDMA, Bis-EMA, TEGDMA Filler: Ba-Al-F-B-Si-glass, Sr-Al-F-Si-glass	0.02–10.0 (mean: 4.2)	68/45	1410000987	Dentsply DeTrey, Konstanz, Germany
x-tra base	Matrix: Bis-EMA, UDMA Filler: Ba-Al-Si-glass, YbF_3, fumed SiO_2	0.05–7.0 (mean: 3.5)	75/60	1419657	VOCO, Cuxhaven, Germany

*ORMOCERS are inorganic-organic hybrid polymers functionalized with methacrylate groups for radical-based cross-linking developed by the Fraunhofer Institute (Würzburg, Germany)

Bis-GMA: bisphenol-A-glycidyldimethacrylate; Bis-EMA: ethoxylated bisphenol-A-dimethacrylate; TEGDMA: triethylene glycol dimethacrylate; UDMA: urethane dimethacrylate

Table 2 Mean values (± standard deviation) of linear shrinkage, shrinkage force, maximum force rate (R_{max}), time to achieve maximum force rate (t_{Rmax}), and degree of conversion of the tested composite materials

Material	Linear shrinkage (%)	Shrinkage force (N)	R_{max} (N/s)	t_{Rmax} (s)	Degree of conversion (%)
Esthet X HD	1.96 (0.16) **C**	21.44 (0.74) **C**	3.96 (1.63) **ABC**	4.26 (0.55) **A**	52.82 (2.89) **C**
Esthet X flow	3.87 (0.09) **A**	35.41 (1.04) **A**	6.62 (2.64) **A**	5.12 (0.79) **A**	60.63 (1.30) **B**
Bulk Ormocer	1.08 (0.03) **E**	12.00 (0.45) **E**	1.94 (0.37) **C**	4.52 (0.72) **A**	56.09 (3.89) **C**
SonicFill	1.44 (0.08) **D**	16.68 (1.24) **D**	4.10 (1.28) **ABC**	4.32 (0.48) **A**	71.82 (2.35) **A**
SDR	2.60 (0.06) **B**	21.47 (1.15) **C**	3.42 (0.38) **BC**	4.32 (0.64) **A**	69.02 (0.48) **A**
x-tra base	2.65 (0.12) **B**	24.83 (1.79) **B**	6.27 (1.79) **AB**	5.46 (0.79) **A**	53.58 (0.94) **C**

Within each column, mean values followed by same capital letters are not significantly different at the 0.05 level (Tukey's HSD post-hoc test)

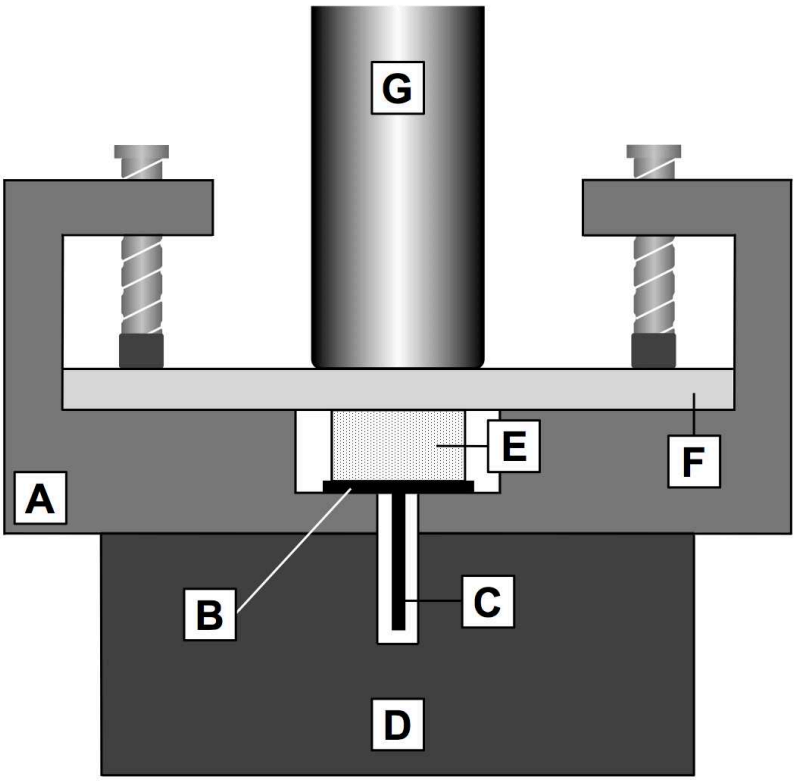

Fig. 1 Diagram of the measuring device (linometer) for linear shrinkage

A: metal frame; B: aluminum platelet; C: diaphragm; D: infrared measuring sensor;

E: composite specimen; F: glass plate; G: light curing unit

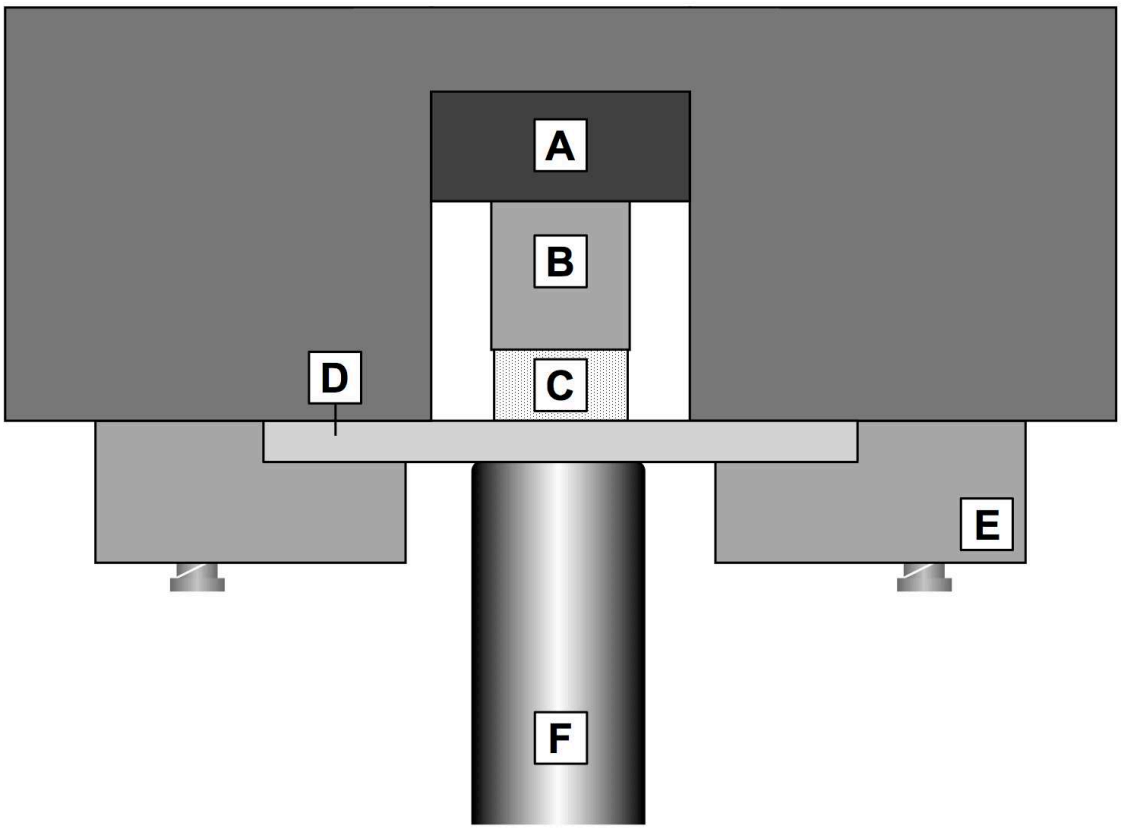

Fig. 2 Diagram of the measuring device for shrinkage force

A: load cell; B: metal cylinder; C: composite specimen; D: glass plate; E: holder of glass plate; F: light curing unit

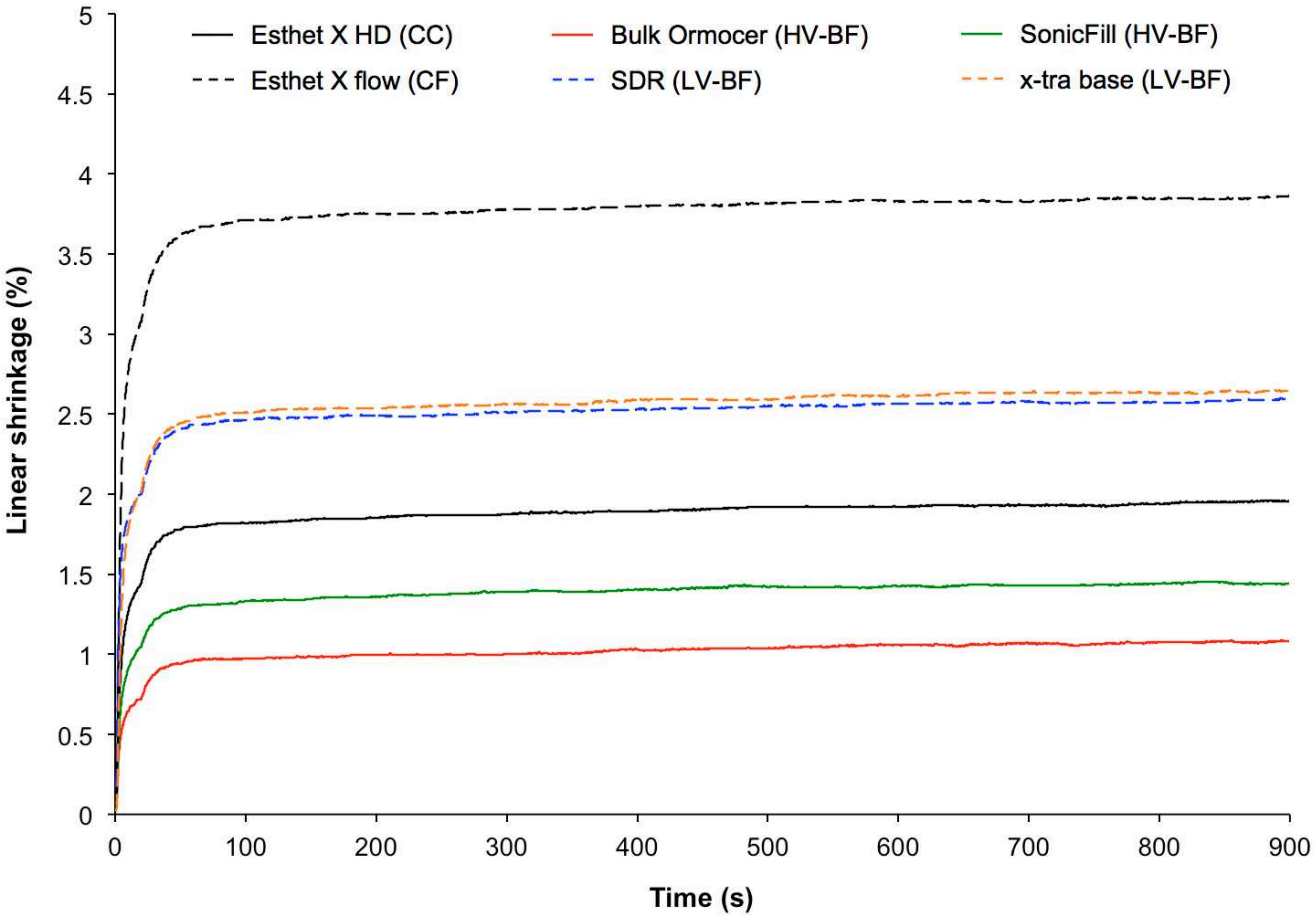

Fig. 3 Mean linear shrinkage curves of the composite materials as a function of time

CC: conventional composite; CF: conventional flowable composite;

HV-BF: high-viscosity bulk-fill composite; LV-BF: low-viscosity bulk-fill composite

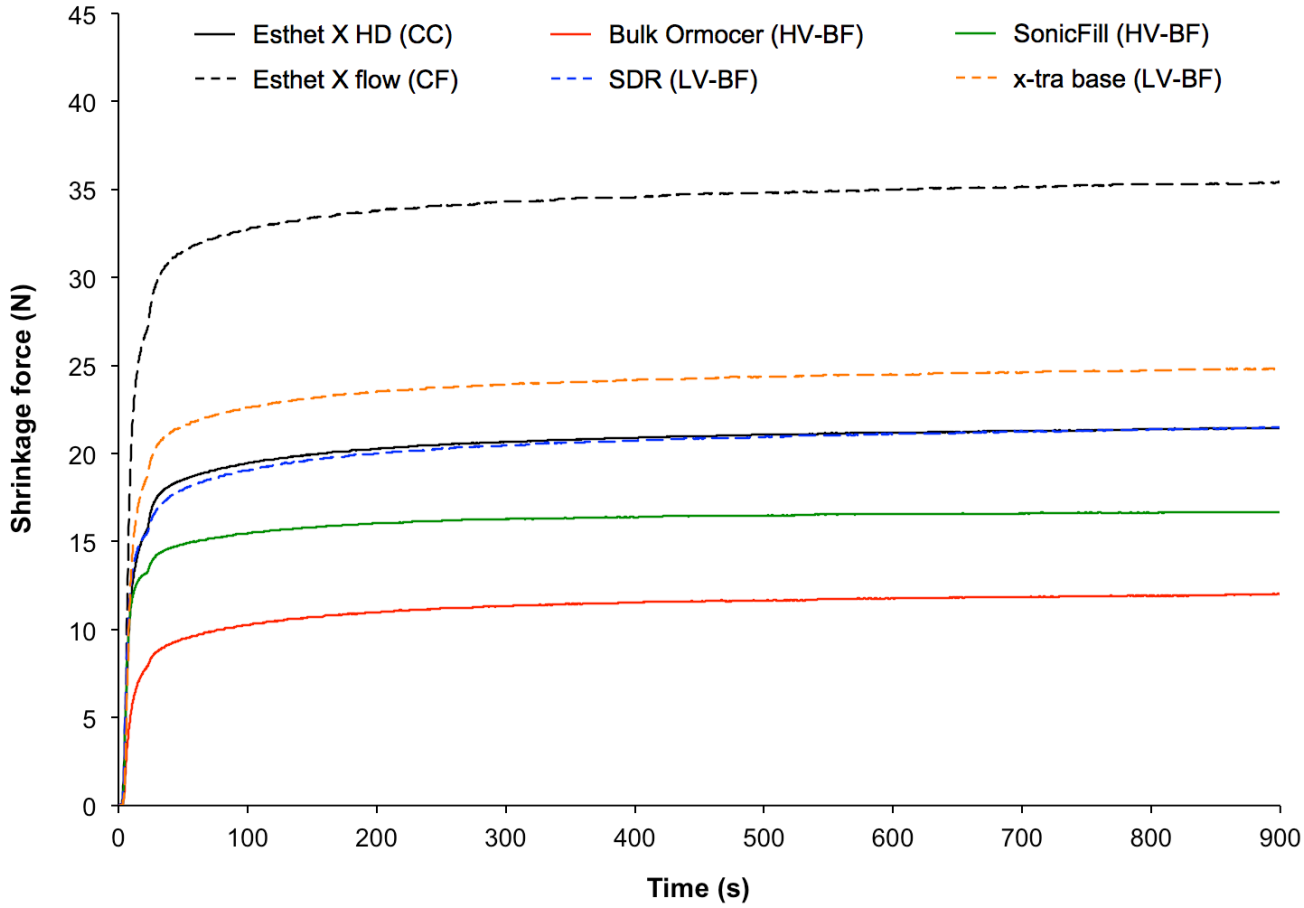

Fig. 4 Mean shrinkage force curves of the composite materials as a function of time

CC: conventional composite; CF: conventional flowable composite;

HV-BF: high-viscosity bulk-fill composite; LV-BF: low-viscosity bulk-fill composite